# INTERMEDIATE REGIME OF LIGHT DIFFRACTION IN CRYSTALS WITH STRONG ELASTIC ANISOTROPY

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**Abstract**
A quantitative model is presented for the evaluation of mismatches and intensities of light diffracted by ultrasound propagating in an acoustically anisotropic medium at normal light incidence. An example is provided in case of ultrasonic light diffraction in $TeO_2$ where the angle between the direction of sound propagation and the acoustic energy flow is 74°.

**Introduction**
Recent progress in acousto-optics (AO) to a great extent is due to the application of materials with an unique combination of physical properties. For example, the crystalline materials paratellurite $TeO_2$, calomel $Hg_2Cl_2$, mercury bromide $Hg_2Br_2$ and lead bromide $PbBr_2$ are among the undoubted candidates for the applications in novel AO-modulators, deflectors and filters [1-3]. The mentioned media are characterized by a strong dependence of their elastic properties on the direction of acoustic propagation in a crystal. This results in the phenomenon that phase and group velocities of elastic waves in these crystals are separated in space by large walk-off angles $j$ [2], which may be as high as 70° or even more [4-6].

Characteristic features of ultrasonic light diffraction (ULD) in elastically anisotropic media differ from the corresponding interaction in elastically isotropic glasses and liquids and have been examined in the papers [4-6]. However, the analysis was mainly concentrated on the Bragg regime of light and sound interaction [5] and on qualitative considerations of the problem as a whole [4,6], while the quantitative distribution of the optical intensity over positive and negative orders of diffraction in the Raman-Nath (RN) and intermediate regime of diffraction has not yet been evaluated. In this paper, a model is presented for the evaluation of ULD in these regimes of diffraction, restricted however to the case of normal light incidence. As an example the diffraction orders observed at the output of a paratellurite crystal in which ultrasound may propagate with a walk-off angle $y$ = 74° are calculated at a frequency of 20 MHz, which is a typical frequency for the intermediate regime.

**Theory**
If a light wave with wave vector $\boldsymbol{k_0}$ interferes with a sound wave with vector $\boldsymbol{K}$ and width $L$, the amplitudes $C_p$ of the resulting diffracted light waves can be calculated from the system [1,6-8]

$$\frac{dC_p}{dx} = \frac{v}{2L}\left[C_{p-1}\exp(i\Delta k_{p-1}x) - C_{p+1}\exp(-i\Delta k_p x)\right] \qquad (1)$$

$$C_p(0) = d_{p,0}$$

where $x$ is a direction orthogonal to the borders of the sound column, $d_{p,0}$ represents Kronecker delta and $v = k|\Delta n|L$ is the RN-parameter, $|\Delta n|$ being the maximum variation of the refractive index $n$ resulting from the sound pressure. The amplitude $C_p$ of the diffraction order $p$ depends on the amplitudes $C_{p-1}$ and $C_{p+1}$ of resp. the preceding and succeeding order. This reflects the fact that during the process of light scattering by sound optical energy is exchanged among neighbouring orders. The efficiency of the optical energy exchange is determined not only by the RN-parameter $v$ but also by the mismatch parameters $\Delta k_{p-1}$ and $\Delta k_p$. In general $\Delta k_p$ represents the magnitude of the mismatch vector $\mathrm{D}\boldsymbol{k_p} = \Delta k_p \boldsymbol{u}$ where $\boldsymbol{k_p}$ refers to the wave vector of the $p$-th order diffracted light wave and $\boldsymbol{u}$ is a unit vector directed orthogonal to the border of the acoustic column. $\mathrm{D}\boldsymbol{k_p}$ essentially describes the phase mismatch which may occur during the process of ULD from order $p$ to $p$+1 [1, 4-6].

It should be noted that the system of coupled differential equations (1) is applicable to describe the process of ULD in isotropic media as well as in media with elastic anisotropy and at both normal and oblique light incidence. The specific conditions of the light/sound interaction are entirely enclosed in the mismatch parameters $\Delta k_p$. Not only $\Delta k_p$ depends on the angle of incidence, i.e. the angle between the wave vectors $\boldsymbol{k}$ and $\boldsymbol{K}$, but it is also affected by the nature of the medium where the AO-interaction takes place. A medium with elastic anisotropy will result in different mismatch vectors with respect to isotropic media. Below we shall first present the AO-interaction in an acoustically isotropic medium. Next we shall consider the consequences of the presence of elastic anisotropy. In both cases, the analysis is restricted to normal light incidence (i.e. parallel to the ultrasonic wave fronts).

*ULD in an isotropic medium*
Fig. (1) shows a scheme of the set-up of ULD at normal incidence in an elastically isotropic medium and the corresponding wave vector diagram of the AO-interaction. From the diagram it follows that

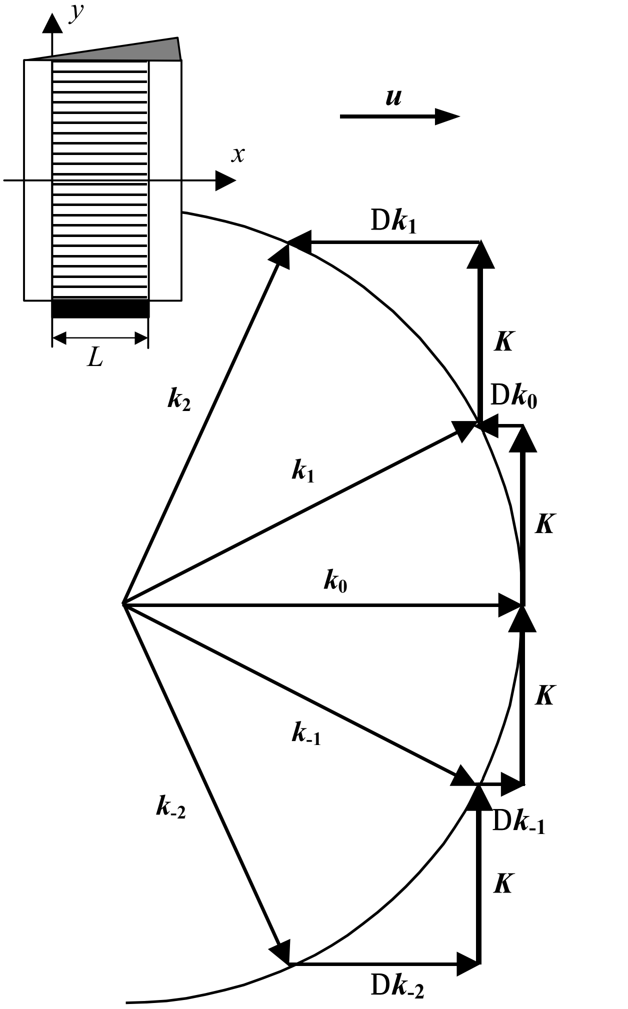


Figure 1: Set-up and wave vector diagram of AO-interaction in an isotropic medium.

$$\boldsymbol{k_p} + \boldsymbol{K} + \mathrm{D}\boldsymbol{k_p} = \boldsymbol{k_{p+1}} \quad (p = 0, \pm1, \pm2, \ldots) \tag{2}$$

where the mismatch vectors D$\boldsymbol{k_p}$ are parallel to the $x$-direction, i.e. the direction of the incident optical wave vector. Upon projecting the vector relationship (2) on the $x$- and $y$-axis, eliminating the angles of diffraction and taking into account the orientation of the unit vector $\boldsymbol{u}$, one can find

$$\Delta k_p = -\sqrt{k^2 - p^2 K^2} + \sqrt{k^2 - (p+1)^2 K^2} \tag{3}$$

where $k = |\mathbf{k_p}|$, $p = 0, \pm1, \pm2, \ldots$ and $K = |\mathbf{K}|$. Note that (3) is negative for $p \geq 0$ and positive for $p < 0$. It is readily verified that the mismatch vectors are equal in pairs but opposite of sign, i.e.

$$\Delta k_0 = -\Delta k_{-1}, \; \Delta k_1 = -\Delta k_{-2}, \text{ etc.} \tag{4}$$

Recalling the mathematical approximations

$$\frac{1}{a+z} \approx \frac{1}{a} - \frac{z}{a^2} \quad , \quad \sqrt{a^2 - z^2} \approx a - \frac{z^2}{2a} \tag{5}$$

valid if $|z| << a$ ($a > 0$), one can deduce from (3)

$$\Delta k_p \approx -(2p+1)\frac{K^2}{2k} = -(2p+1)\frac{Q}{2L} \tag{6}$$

where $Q = K^2L/k$ represents the Klein-Cook parameter and which is a useful relationship valid whenever

$$pK << k \text{ or } pK/k << 1 \tag{7}$$

for all orders $p$ involved in the diffraction process. The relationships (3) and (6) reveal the dependency of the mismatch vectors on the ultrasonic frequency. It should be recalled that in general $\Delta k_p$ also depends on the angle of incidence.

If the ULD takes place under circumstances where condition (7) is reliable, system (1) is formally equivalent to the well-known original Raman-Nath system

$$\frac{df_p}{dx} - \frac{v}{2L}(f_{p-1} - f_{p+1}) = ip^2 \frac{Q}{2L} f_p \ , \qquad f_p(0) = d_{p,0} \tag{8}$$

which is a linear system with constant coefficients. It suffices to perform the substitution

$$C_p(x) = f_p(x)\exp(-ip^2 \frac{Q}{2L} x) \tag{9}$$

to see the equivalency. A well-known property of system (8) is that its solution satisfies the symmetry relation

$$f_{-p} = (-1)^p f_p \tag{10}$$

which means that the intensities $I_p = |f_p|^2$ of opposite order are identical and that phases of opposite even orders are equal and shifted by π in case of odd orders. Taking into account (4), it is readily verified that the symmetry property (10) also is fulfilled by the solution of the non-approximated system (1), i.e.

$$C_{-p} = (-1)^p C_p \tag{11}$$

In the next section we will see that the symmetry is affected in the presence of elastic anisotropy.

*ULD in an acoustically anisotropic medium*

In a medium with acoustic anisotropy, the direction of propagation of an acoustic wave in general is not the same as that of the energy flow (Poynting vector). The so-called *obliquity angle* or *walk-off angle* $y$, i.e. the angle between the directions of phase and group velocity, in some crystals may exceed 70° and it may be positive or negative. Fig. (2a) shows the geometry in the case of a positive walk-off angle ($y > 0$) while Fig. (2b) represents the case $y < 0$. Figure (2c) shows the wave vector diagram in case $y > 0$. It should be noted that the walk-off angle $y$ also appears as the angle between the direction of the incident light wave vector and the mismatch vectors $\mathrm{D}\boldsymbol{k}_p = \Delta k_p \boldsymbol{u}$ where $\boldsymbol{u}$ represents a unit vector oriented as in Fig. (2). By projection of the vector sum (2) on both the *x*- and *y*-direction and elimination of the diffraction angles, one can deduce that

$$\Delta k_p = K\sin y - \sqrt{(k\cos y - pK\sin y)^2 - p^2K^2} + \sqrt{[k\cos y - (p+1)K\sin y]^2 - (p+1)^2K^2} \qquad (12)$$

The formula is valid for both positive and negative walk-off angles $y$. In case $y = 0$, (12) reduces to (3). It should be noted that, due to the dependency of the mismatch vectors on the walk-off angle, the symmetry with respect to diffraction orders of opposite sign in general is lost. In particular, putting $p = 0$ and $p = -1$, one can find for the mismatch parameters in which the zero-order diffracted light beam is involved that

$$\Delta k_0 = -(k\cos y - K\sin y) + \sqrt{(k\cos y - K\sin y)^2 - K^2} \qquad (14)$$

$$\Delta k_{-1} = k\cos y + K\sin y - \sqrt{(k\cos y + K\sin y)^2 - K^2}$$

where still $\Delta k_0 < 0$ and $\Delta k_{-1} > 0$ but the mismatches have different absolute values.

Again applying the formulae (5), one can find the following approximated expression for the mismatch parameters:

$$\Delta k_p \approx -\frac{K^2}{2k\cos y}\left[2p + 1 + (3p^2 + 3p + 1)\frac{K}{k}\tan y\right] \qquad (15)$$

which again is valid whenever $pK << k$. If $y \neq 0$, the 2[nd] term between the brackets is responsible for the broken symmetry, e.g.

$$\Delta k_0 \approx -\frac{K^2}{2k\cos y}\left[1 + \frac{K}{k}\tan y\right] \qquad (16)$$

$$\Delta k_{-1} \approx \frac{K^2}{2k\cos y}\left[1 - \frac{K}{k}\tan y\right]$$

Under the conditions where the approximation is reliable, system (1) may be further simplified to a linear system with constant coefficients. Rewriting (15) as

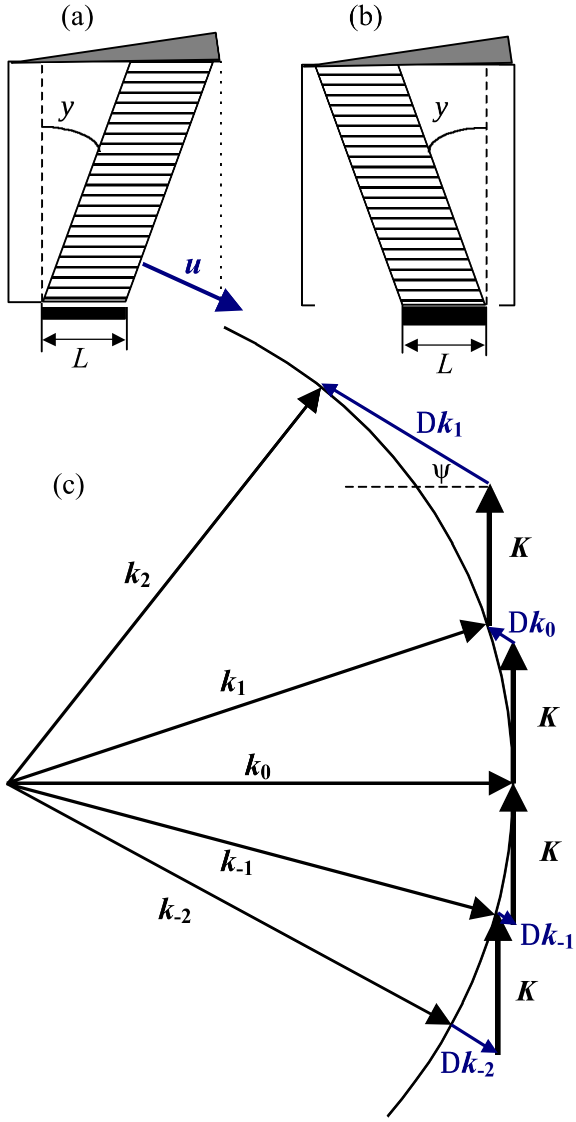


Figure 2: ULD in an elastically anisotropic medium at light incidence parallel to the sound wave fronts. (a) positive obliquity; (b) negative obliquity; (c) wave vector diagram corresponding to $y > 0$.

$$\Delta k_p \approx (2p + 1)a_1 + (3p^2 + 3p + 1)a_2 \qquad (17)$$

where

$$a_1 = -\frac{K^2}{2k\cos y} = -\frac{Q}{2L\cos y} \qquad (18)$$

$$a_2 = -\frac{K^3\tan y}{2k^2\cos y} = \frac{K\tan y}{k}a_1 \qquad (19)$$

and performing the substitution of

$$C_p(x) = f_p(x)\exp(ip^2 a_1 x)\exp(ip^3 a_2 x) \qquad (20)$$

into (1), the following system is obtained:

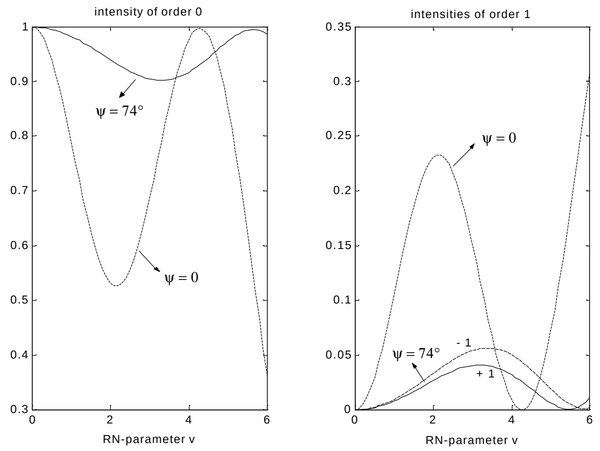


Figure 3: ULD at 20 MHz in $TeO_2$ in case of normal light incidence and $y$ = 74°. Intensities of order 0 and ±1 versus *v*. Dashed lines refer to the case $y$ = 0.

$$\frac{df_p}{dx} - \frac{v}{2L}(f_{p-1} - f_{p+1}) = -ip^2(a_1 + pa_2)f_p \qquad (21)$$

$$f_p(0) = d_{p,0}$$

This system can be considered as an extension of the original Raman-Nath system (8) to a medium with elastic anisotropy. The anisotropy is reflected by the phase term $\alpha_2$ and the appearance of the walk-off angle $y$. In an acoustically isotropic medium, $y$ = 0 and (21) reduces to (8). In the RN-regime, the right member tends to zero and symmetry is restored.

## Example

To illustrate the asymmetry in general appearing as a result of the acoustic anisotropy, we have selected $TeO_2$ (paratellurite) as an AO-medium. This crystalline material, which is commonly used in modern acousto-optics, exhibits a very strong dependence of the acoustic properties on the direction of sound propagation. At a specific crystal cut the walk-off angle $y$ is as large as 74° in the case of a shear ultrasonic mode [6]. The refractive index of the medium is $n$ = 2.41, the sound velocity $V$ = 980 m/s and the effective transducer width $L$ = 0.8 cm. An optical wavelength of 0.633 $\mu$m and a sound frequency of 20 MHz are further assumed. Under these circumstances, the Klein-Cook parameter is $Q$ = 5.5 and hence the ULD takes place in the so-called *intermediate* diffraction regime, i.e. the regime between Raman-Nath and Bragg regime. Figure (3) shows the light intensities of the orders 0 and ±1 against *v*. As a reference, the light intensities in the isotropic case $y$ = 0 are included. All curves can be calculated from either system (1) or (21) as the condition (7) is fulfilled ($K/k$ = 0.0054). The strong influence of the walk-off angle is obvious: the intensity levels of both the zero and first orders are seriously affected by the acoustic anisotropy and also the asymmetry between orders of opposite sign appears. Due to the positive obliquity, the negative first order of diffraction is subject to a smaller mismatch than the positive one and hence is favoured, at least within the considered (realistic) range of $v$ .

## Conclusion

In contrast with ULD in an isotropic medium, it is shown that in a medium with elastic anisotropy and at normal light incidence, asymmetry appears in the light intensities of opposite diffraction orders. The asymmetry increases with the angle between the direction of acoustic propagation and the energy flow. It is also demonstrated that the level of the diffracted light intensities may be seriously affected by the anisotropy. Inversely, both effects can be related to changes in the obliquity angle $y$ and hence can provide information about the acoustic anisotropy of the medium. The theoretical model presented allows the calculation of the diffracted light intensities at normal light incidence, including the mismatches introduced by the acoustic anisotropy.

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